Stable In-reactor Performances at Low Temperature of U-10wt.%Mo Dispersion Fuel Containing Centrifugally Atomized Powder

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ABSTRACT

In order to examine the in-reactor performances of very-high-density dispersion fuels for high flux performance research reactors, U-10wt.%Mo microplates containing centrifugally atomized powder have been irradiated at low temperature. The U-10wt.%Mo dispersion fuels show stable in-reactor irradiation behaviors even at high burn-up, similar to U_3Si_2 dispersion fuels. The atomized U-10wt.%Mo fuel particles have a fine, uniform fission gas bubble size distribution. Moreover, only one third areas of the atomized fuel cross-sections at 70at.% burn-up show fission gas bubble-free zones. This appears the result of segregation into high Mo and low Mo.

1. Introduction

The conversion from high enriched uranium (HEU) to low enriched uranium (LEU) for use in research reactor fuel requires a large increase of uranium per unit volume to compensate for the reduction in enrichment. The relatively high- density compound U_3Si_2 , with a uranium density of 11.6 g-U cm^{-3}, was found to possess a very stable irradiation behavior; however, fabricability limits do not allow fuel element loadings higher than 6 g-U cm^{-3} [1-5]. Therefore, very-high-density fuels having U-loadings up to 8~9 g-U cm^{-3} require both a very dense fuel dispersant (>15 g-U/cm^3) and a very high volume loading in the dispersant (>50 vol.%). Uranium alloys including small amounts of alloying elements can be considered as fuel dispersants [6-7]. The only uranium compounds having such densities are the U_6X compounds, such as U_6Fe and U_6Mn, which have been shown to perform poorly under irradiation [8]. However, alloys of uranium and some transition elements that maintain uranium in the metastable γ (cubic) phase have shown good irradiation performance in bulk form to intermediate burn-up under fast reactor conditions. Sufficiently small amounts of one such alloying element, molybdenum, had been shown to stabilize γ-U and at the same time yield a high uranium density [9].

The two key issues are the reaction of the fuel alloy with the aluminum matrix and the irradiation behavior of the dispersion. The former issue is important because excessive reaction will consume the matrix aluminum and possibly a significant amount of the aluminum-alloy cladding. The latter issue relates principally to the behavior of the fission gas in the fuel. If the mobility of the fission gas is small enough, it will be contained in small bubbles that do not interlink, as shown for U_3Si_2 [4-5]. Such a fuel will exhibit a steady but stable increase in volume during irradiation. On the other hand, if the fission gas is very mobile, some bubbles grow preferentially, becoming large and interlinking with adjacent bubbles, as shown for U_3Si [10-11]. If the volume loading of such fuel particles is high enough that a significant number of particles are touching, the fission gas bubbles can interlink across many particles and lead to unstable, rapid (breakaway) swelling.

Early irradiation experiments with uranium alloys showed the promise of acceptable irradiation behavior, if these alloys could be maintained in their cubic γ-U crystal structure [12]. If centrifugally atomized U-Mo powder can retain this gamma uranium phase during fuel element fabrication and irradiation, and if it is compatible with aluminum which forms the matrix of dispersion fuels, the uranium alloy would be a prime candidate for dispersion fuel for research reactors. Very-high-density atomized U-10wt.%Mo powder prepared by centrifugal atomization retained the isotropic high temperature γ-U phase [13]. Moreover, the gamma phase did not decompose into the equilibrium γ-U and U_2Mo two phase structure in U-10wt.%Mo alloy after annealing of up to 100 hours at 400°C. In addition, the U-10wt.%Mo particles dispersed in aluminum did not show significant dimensional changes after annealing up to 2,000 hours at 400°C, and interdiffusion between U-10wt.%Mo and aluminum was found to be minimal [14]. In this study, in order to characterize the in-reactor performances of the atomized U-10wt.%Mo dispersion fuels, the U-10wt.%Mo microplates have been irradiated to approximately 40at.% and 70at.% burn-up at low temperature.

2. Experimental procedure

Low enriched uranium lumps (99.9 pct pure) and molybdenum buttons (99.7 % pct pure) were used for the preparation of the U-10wt.%Mo powders by rotating-disk centrifugal atomization [13]. Dispersion fuel meats with a nominal volume fraction of 25% were prepared by blending the U-10wt.%Mo and aluminum powder and by rolling the blended powders at a working temperature of about 485°C.

The microplate fuel samples were fabricated with external dimensions of 76 mm x 22 mm x 1.3 mm in aluminum cladding. The fuel zone is elliptical in shape with major and minor axes of approximately 51 mm and 9.5 mm, respectively; the fuel zone thickness is nominally 0.5 mm. U-10wt.%Mo microplates, which were irradiated at an average fuel center temperature of 65°C, were discharged after 94 effective full-power days (EFPDs) of irradiation, and then discharged after 232 EFPDs of irradiation, achieving (calculated) ^{235}U burnups of 40at.% and 70at.%, independently. Thereafter, post-irradiation examinations of the microplates are performed, primarily using a scanning electron microscope.

3. Experimental results

Fig. 1 shows scanning electron micrographs of fracture surfaces in the fuel meat of atomized U-10wt.%Mo microplate at 40at.% burn-up. The fuel-matrix interaction layer of the atomized spherical particles is very uniform and in the range of ~2 μm in thickness at 40at.% burn-up. Fission gas bubbles generally appear to be concentrated on the primary grain boundaries almost exclusively. Fig. 2 shows apparent fission gas bubble size distributions at 40at.% burn-up measured on fracture surfaces. The maximum bubble diameter of atomized fuel is approximately 0.5 μm, while the average bubble diameter is approximately 0.2 μm.

Scanning electron micrographs of fracture surfaces in the fuel meats of U-10wt.%Mo microplates at 70at.% burn-up are shown in Fig. 3. There are fission gas bubble-free zones of ~35% in area fraction and 5~10 μm in zone in the atomized particles. Gas bubble-free zones, not entering the stage of bubble formation, are located primarily around the perimeter rather than toward the center of the atomized particle. Small bubble-free zones are sometimes formed in bubble-rich zones in places. Back-scattered scanning electron image of an atomized U-10wt.%Mo microplate at 70at.% burn-up is also shown in Fig. 4-(a). The average size of bubble-free islands of 3~10 μm is almost similar to the homogenized grain size of 5~10 μm. Fission gas bubbles are associated with a granular microstructure that appears to have nucleated along grain boundaries and then grow continuously toward the center of grains, leaving many bubble-free zones within grain. A somewhat different picture is shown in Fig. 4-(b), when the scanning electron microscopy samples are polished. The granular appearance of the fracture surfaces, which indicated grain refinements, is no longer visible and a more representative gas bubble morphology can now be described. The bubbles, as well as the grain refined microstructure, with the lower Mo segregated γ phase that formed during solidification in the atomization process [13]. The bubble-free areas, on the other hand, are the high Mo γ phase fraction that apparently did not recrystallize (formed fine grains) at 70at.% burnup and did not develop visible gas bubbles.

Fig. 5 shows apparent fission gas bubble size distributions for two U-10wt.%Mo microplates at 70at.% burn-up measured on fracture surfaces. The bubble size and the population density increase greatly, as the burn-up of the microplates increases from 40at.% to 70at.%, and the maximum bubble diameter of both atomized and ground fuel particles is approximately 0.7 μm and 1.0 μm, respectively, and the average bubble diameter is approximately 0.2 μm and 0.3 μm, respectively. The population density of the atomized fuel powder is about 1.4×10^{12} m^{-2}. Whereas, the ground powder has a prominent bimodal bubble size distribution and a higher population density of about 2.6×10^{12} m^{-2}, two times as many as the population density of the atomized powder. This is due to the combination of; the presence of the second peak in the bubble distribution and the absence of bubble-free areas in the ground fuel.

4. Discussion

The in-reactor performance of U-10wt.%Mo microplates, irrespective of powder kind, shows a stable irradiation behavior even at high burn-up, similar to that of U_3Si_2 [1-5]. The U-10wt.%Mo dispersion fuels do not show any indication of breakaway swelling [15]. A U-10wt.%Mo alloy can exist in two states: the heterogeneous state appears to be stable at room temperature; a homogeneous solution of Mo in γ-uranium appears to be stable above 570°C. It can crystallize either in the form of a supersaturated α solid solution or in the form of γ' phase. An as-cast U-10wt.%Mo ingot was heat-treated in vacuum for 100 hrs at 900• to ensure compositional homogeneity, and then quenched to form a metastable γ-U phase [16]. The mechanically ground powder prepared from the as-cast ingot contained some α-U centering grain boundaries, which must have come from the partial decomposition of the metastable γ-U phase during hot rolling, performed at 500°C for 2 hrs, in contrast to the atomized U-10wt.%Mo powder. There is no evidence of interlinking of the relatively uniformly distributed fission gas bubbles, even in the ground powder. It indicates that there is no obvious evidence of a two-phase microstructure in any of the irradiated microplates, irrespective of powder kind [16]. Therefore it seems that the partial decomposition of the γ-U phase in the ground powder which occurs during fabrication is probably reversed early during irradiation. It has been well established that a transition (α-U + $U_2Mo \rightarrow$ γ solid solution) is caused by irradiation, even when the mean temperature of the specimen does not exceed 100~150°C. Such an effect has been reported in the literature as owing to fission-spike mixing at high fission rates. The removal of heat from the neighborhood of such a thermal spike is accomplished in a very short period of time. As a result, the formation of a spontaneous crystallization centre in the thermal spike area seems improbable and the crystallizing volume acquires the structure of one of its neighboring lattices. Hence, as the diffusion and alignment of the average molybdenum composition in the U-10wt.%Mo particles develop further in spite of an average fuel center temperature of 65°C, there takes place a transition, into the γ phase.

The atomized U-10wt.%Mo dispersion fuel has a finer, more uniform fission gas bubble size distribution and lower population density of gas bubbles than the mechanically ground dispersion fuel. The possible reasons are supposed as follows. The atomized particles do not have prominent fabrication damage. Whereas, the ground particles have severe deformation damage with high dislocation density formed during mechanical comminution process. During rolling or irradiation of the ground U-10wt.%Mo dispersion fuels, the dislocations interact and tend to cluster into arrangements of high dislocation density that are separated by regions with relatively low dislocation density [17-19]. The clustering of dislocations is a general observation in deformed polycrystalline alloys [19-21]; typical dislocation configurations are dislocation tangles, two-dimensional dislocation boundaries (or walls), and three-dimensional dislocation cell structures. The different dislocation configurations derive from energy minimization, where glide-dislocation configurations increasingly approach the minimum energy per unit length of dislocation line as dislocation density - and hence interaction between dislocations - increases. Strain-free blocks within a crystal are formed by the spatial rearrangement of dislocation into lower-energy arrays by polygonization [22-24]. Recrystallization has generally occurred in all of the alloys, resulting in an average grain size of submicron. It is promoted by increasing amounts of cold work, as the activation energy for recrystallization is a function of the amount of deformation. The heavily deformed areas of the ground U-10wt.%Mo have highly stored strain

energy with a high degree of deformation, that is, initial dislocation density. They tend to nucleate and grow into subgrains around grain boundaries during hot rolling or irradiation. Hence, the onset of grain subdivision in the ground U-10wt.%Mo occurs at a lower burn-up vis-á-vis the atomized powder. The ground U-10wt.%Mo at 70at.% burn-up has a granular appearance all over the particles, suggesting a grain refinement of about 0.8μm in subgrain size similar to coarse fission gas bubble size. However, the atomized powder at 70at.% burn-up shows a partial granular appearance and bubble-free zones. It indicates that a severe grain subdivision by recrystallization did not yet occur in some regions of the atomized particles.

Irradiation-induced recrystallization and enhanced bubble growth on the newly formed grain boundaries were also observed for UO_2 power reactor fuels. Recrystallization and intergranular bubble growth have been definitively confirmed for UO_2 fuels. "Subdivision" of the original grains has been observed in high burn-up uranium dioxide. The peripheral region of LWR fuel pellets reveals an increasingly porous microstructure with burn-up [25-29]. Observation of this "rim effect" showed that an extremely fine-grained structure formed by subdivision of the original fuel grain was associated with the porous microstructure. The large fission gas bubbles generally appear to be forming at the subgrain boundaries, as the recrystallized boundaries around grain boundaries have very high fission gas mobility. Upon grain subdivision in U-10wt.%Mo fuel particles, fission gas atom diffuses from the bulk to the grain faces, where it accumulates in gas bubbles that grow until grain-face saturation occurs. The coarse bubbles occurring at a bubble diameter larger than 0.25 μm visible gas bubbles shown in Fig. 5 are associated with grain-corner bubbles formed by the intersection of grain edges within the subgrain boundary structure. Fission gas that collects along grain edges vents upon intersection to these ″dead-end″ nodes grow as they continue to collect additional gas. The large bubbles of U-10wt.%Mo fuel particles in the grain subdivision region are due to the accumulated gas in the ″dead-end″ nodes. Moreover, in the case of where fabrication-induced damage leads to an earlier onset irradiation-induced recrystallization, the higher gas bubble population will occur at a larger bubble size. This is because of the shorter time that bubbles have to accumulate and grow on the subgrain boundaries. Subsequent to irradiation-induced recrystallization, the growth rate of gas bubbles on the recrystallized grain surface increases. Fission-gas bubbles nucleate at the newly formed boundaries and then grow at an accelerated rate relative to that of fission-gas bubbles in the bulk material. They accelerate the growth rate of gas bubbles of the ground U-10wt.%Mo particles at a lower burn-up vis-á-vis atomized particles. The fine bubbles occurring at a bubble diameter smaller than 0.25 μm shown in Fig. 5 are associated with grain boundary bubbles. In contrast, the coarse bubbles of the atomized U-10wt.%Mo at 70at.% burn-up are formed upon grain subdivision. About one-third of the area of bubble-free zones in the atomized fuel cross-sections at 70at.% burn-up, which appears to be associated with the segregated higher Mo fraction of the γ phase, indicates that a considerable amount of the fuel particle does not start grain refinement.

5. Conclusions

In order to examine the in-reactor behavior of very-high-density dispersion fuels for high flux performance research reactors, U-10wt.%Mo dispersion fuels containing centrifugally atomized powder have been irradiated at low temperature.

1) The U-10wt.%Mo dispersion fuels show very stable in-reactor irradiation behavior, indicating the maintenance of the metastable cubic γ-U phase .
2) The atomized U-10wt.%Mo fuel particles have an overall finer, more uniform fission gas bubble size distribution and lower population density of gas bubbles relative to the mechanically ground fuel particles.
3) The atomized U-10wt.%Mo fuel particles at 70at.% burn-up show fission gas bubble-free zones, in contrast to the ground fuel cross-sections containing very small fission gas bubble-free zone.
4) The possible reasons for different in-reactor behavior of the atomized powder relative to the ground powder are supposed as follows.
   i) The atomized particles do not have severe deformation damage with high dislocation density formed during mechanical comminution process.
   ii) The grain refinement and the gas bubble formation in the ground particles occurs at a lower burn-up than in the atomized particles.
   iii) The atomized particles contain 2 sub γ-phase fractions as a result of segregation (coring).

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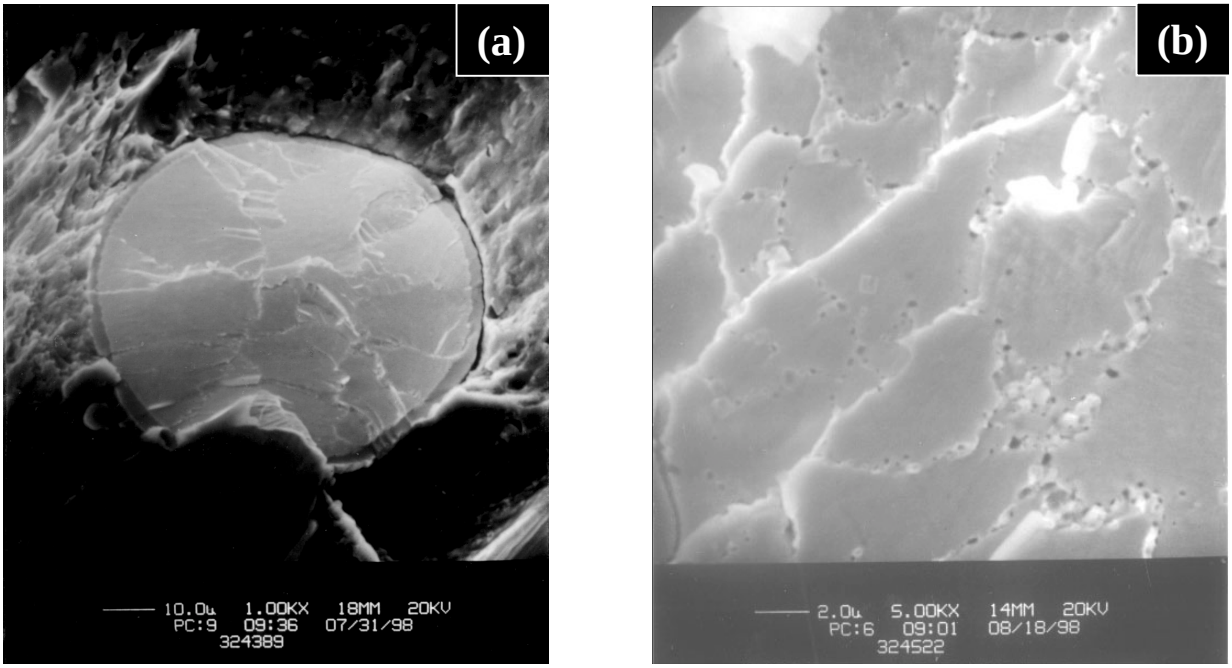


Fig. 1. Scanning electron micrographs of fracture surfaces in the fuel meat of U-10wt.%Mo microplates at 40at.% burn-up; (a) x1,000, (b) x5,000.

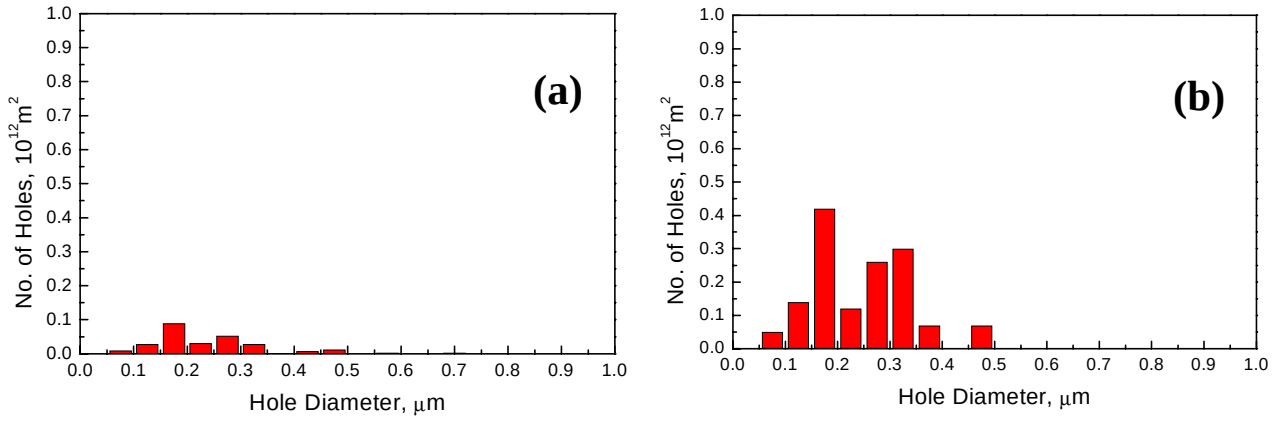


Fig. 2. Apparent fission gas bubble size distributions on fracture surfaces of U-10wt.%Mo microplates at 40at.% burn-up; (a) Atomized fuel powder, (b) Ground fuel powder.

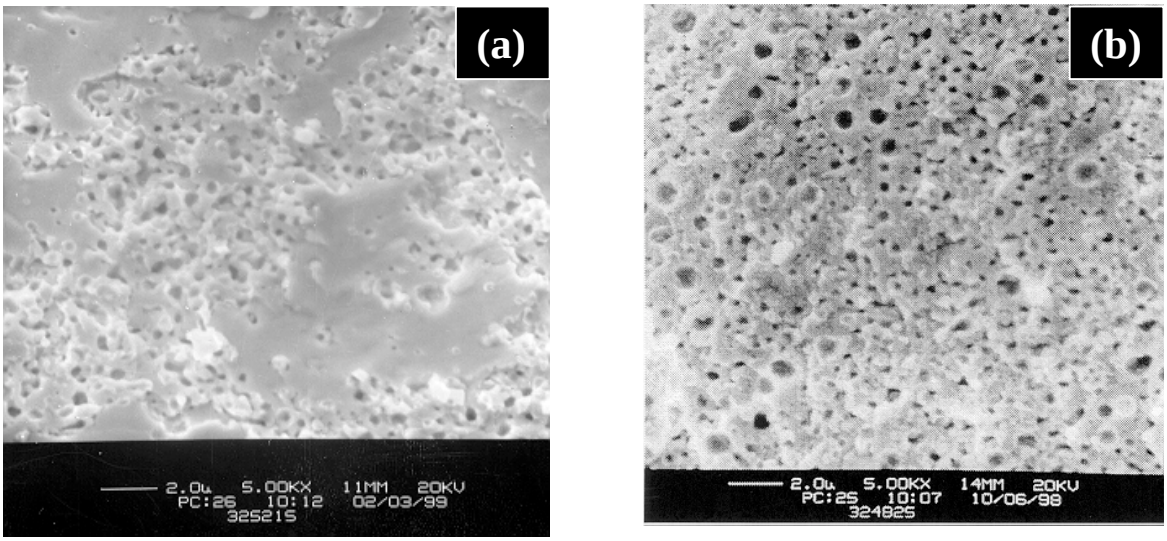


Fig. 3. Scanning electron micrographs for U-10wt.%Mo microplates at 70at.% burn-up; (a) Atomized fuel powder, (b) Ground fuel powder.

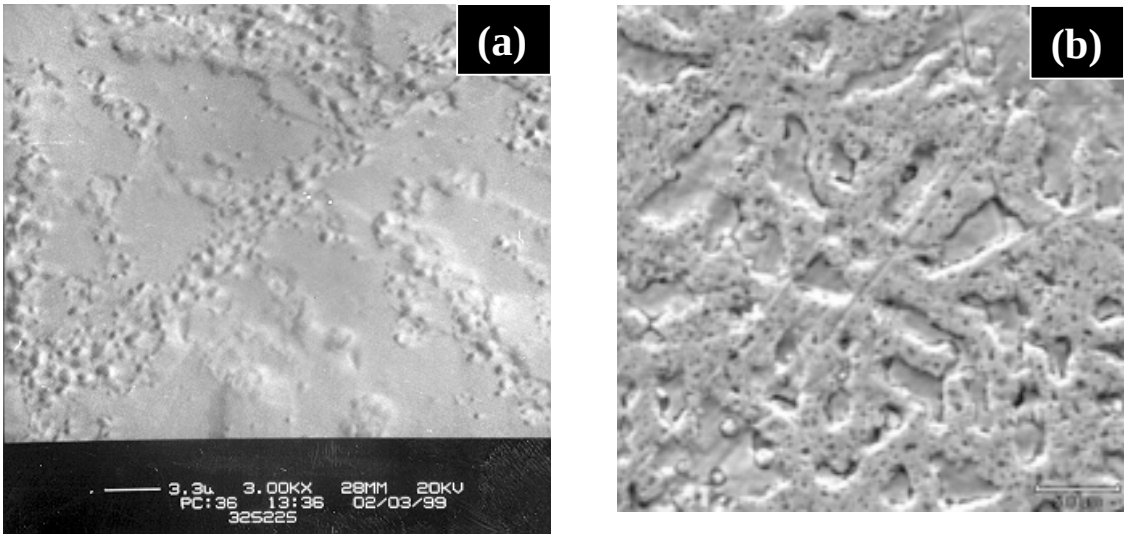
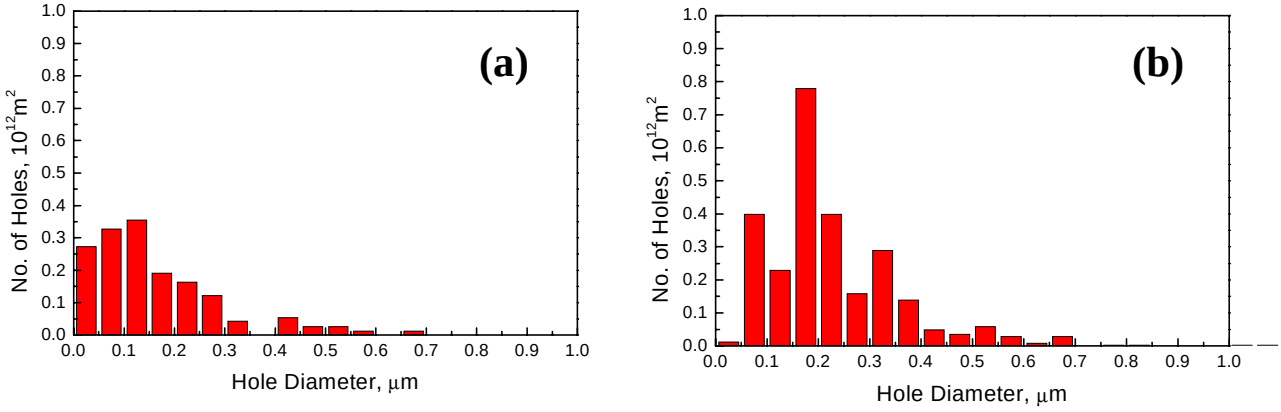


Fig. 4. Back-scattered scanning electron image of a fractured surface (a) and scanning electron image of a polished surface (b) of atomized U-10wt.%Mo microplate at 70at.% burn-up.



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